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The fiftieth anniversary of the first applications of the scanning electron microscope in materials research

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Abstract

The paper summarises the early attempts during the 1930s in Berlin and New York to build scanning electron microscopes (SEMs), and the revival of the subject in 1948 by Charles Oatley at the Engineering Department in Cambridge University in England. The first Cambridge SEM was working in 1951 and after further development was shown in 1956 to be capable of imaging specimens that could not be examined in a conventional transmission electron microscope (TEM). Five of these applications are described. After several more years, the advantages of the SEM for imaging surfaces were finally accepted by most electron microscopists and the first SEM was marketed by the Cambridge Instrument Company in 1965. © 2008 Elsevier B.V. Open access under CC BY-NC-ND license.

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1. Introduction

The scanning electron microscope (SEM) was invented by Max Knoll in 1935, at the Telefunken Company in Berlin, for studying the secondary emission properties of television camera tube targets [1]; four years earlier, he and Ernst Ruska had built the first transmission electron microscope (TEM). Knoll's electron beam scanner had virtually all the features of a SEM – the beam was scanned to produce a 200-line, 50-frames/sec raster, and a cathode-ray tube was deflected in synchronism and modulated in brightness by the specimen current to display a real-time visible image. It employed a single demagnifying lens to focus the beam into a probe diameter of 100 μm with which he demonstrated several important properties of the SEM, with magnifications of up to about 10 times.

The first attempt at building a scanning microscope with a sub-micrometre probe was made in 1937 by Manfred von Ardenne in his private laboratory, also in Berlin. His instrument operated as a scanning transmission electron

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microscope (STEM) using photographic recording, but not as a SEM because of the lack of a suitable detector. The most important results from his research were two papers [2-3] on the design and performance of probe-forming electron optics using magnetic lenses, and his analysis covered the limitations on probe diameter due to lens aberrations, diffraction, and the current in the probe. A few years later he proposed the use of an electron multiplier for a SEM detector and analysed how contrast would be formed from a solid sample [4-5]; this formed the basis of much of the later work by others.

At about the same time, a SEM was being developed at the RCA laboratories in New Jersey under the direction of Vladimir Zworykin, the well-known inventor of television camera tubes, and James Hillier, the Canadian pioneer of the TEM [6]. This instrument incorporated an electrostatic immersion lens as the objective in which the beam electrons were slowed to about 900 eV before hitting the sample. Secondary electrons from the specimen were accelerated back up the column and hit a phosphor screen optically coupled to a photomultiplier. This was developed many years later by Everhart and Thornley [7] into the detector that is still widely used for SEM. In spite of these advanced features the microscope was only a partial success because there was no provision for producing a real-time image and recording took 10 min using a fax machine. The original intention of relying on variations in secondary emission ratio to provide image contrast was not successful because of the poor vacuum. A typical micrograph of etched brass is shown in Fig. 1. After the replica process for imaging surfaces with the TEM was described in 1941 by H. Mahl [8], the SEM was reckoned to be of little importance and RCA discontinued its development and concentrated on the production of TEMs.

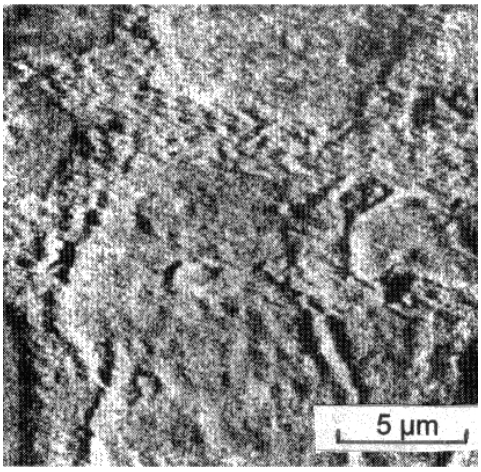

Fig. 1. Micrograph of etched brass produced by the SEM of Zworykin et al.; horizontal field width = 18 µm. [6]

Apart from some experimental work by Léauté [9] in Paris during the war and a theoretical paper on noise by Brachet [10], it appears that for several years there was no further work on SEM anywhere.

2. Charles Oatley

This was the position when in 1945 Charles Oatley was appointed lecturer at the Cambridge University Engineering Department (CUED), and decided that an electrostatically focussed TEM would be a suitable project for a Ph.D. student, K.F. Sander. About a year later Sander abandoned it and instead developed an electron trajectory plotter. The TEM project was not restarted because in the meanwhile V.E. Cosslett had joined the Cavendish Laboratory, Cambridge, to form an electron microscope group and as Oatley wrote later " it was important to avoid trespassing on his ground, but in the outcome this has never caused any difficulty" [11].

A few years afterwards he decided that a SEM would a suitable alternative project. During the war Oatley had been Acting Superintendent of the Radar Research and Development Establishment [12] and was well aware of the advances in electronic technology that might be applied in such an instrument. Another factor was that A.S. Baxter in the Cavendish had developed an electron multiplier with beryllium copper dynodes [13]. This appeared to Oatley to be a suitable detector for a SEM because it could be opened to the atmosphere without damage, and moreover Baxter was willing to lend him one.

3. First Cambridge SEM

Oatley had thus set the scene for this further effort to develop a SEM and as the research student he chose D. McMullan who had had several years experience in radar development and TV projection cathode-ray tubes. What McMullan did not know when he started was that Oatley had been told by electron microscopy experts that an attempt to develop a SEM would be "a complete waste of time", and it was not long before he was also the target of such comments.

In 1948, money was short and virtually everything had to be made in-house; McMullan began by building an electrostatically focussed 40-kV TEM using some of the parts designed by Sander and then converted it to a STEM with Baxter's electron multiplier as the detector. The image was viewed directly on a long- persistence cathode-ray tube with a 1.8-sec frame time, and recorded photographically over a longer period [14]. Consideration was then given to how a SEM image might be obtained. The main lesson that seemed to be apparent from Zworykin's work was that a high energy beam should be used, but it was not clear how image contrast might be formed. Images of surfaces which had been obtained at grazing incidence in the TEM by von Borries [15] and by others, particularly J.W. Menter [16] of the Cambridge Department of Physical Chemistry, pointed the way, and McMullan mounted a specimen of etched aluminium at a relatively large angle to the incident beam (30 degrees), and the high-energy scattered electrons in the forward direction were detected. The image produced gave the now well-known three-dimensional impression as can be seen in Fig. 2 which was a direct view 1.8-s frame period image of etched aluminium and also one recorded for 5 min [17]. The beam energy was 16 keV and the probe diameter about 50 nm. Only a small proportion of the low-energy secondary electrons were collected and, in fact, McMullan erroneously believed that they would be deleterious to the image.

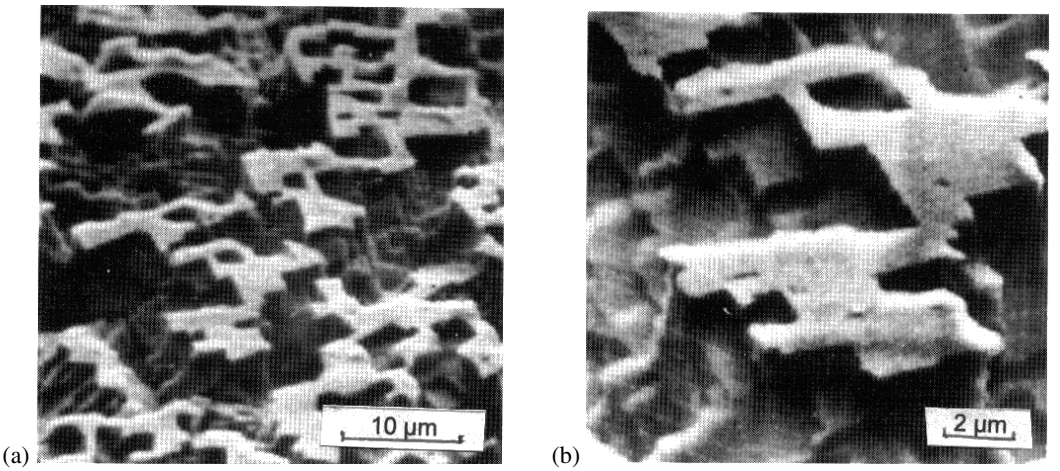

Fig. 2. Cambridge SEM1: first micrographs of etched aluminium. Angle of incidence of 16 keV electrons 25 deg. (a) visible image (1.8-sec scan), beam current 1.5 x 10^{-10} A, horizontal field width = 37µm; (b) 5 min recording, 10^{-13} A, horizontal field width = 15 µm. [17]

Other innovations included an attempt to record an atomic number contrast image with a beam at normal incidence, double-deflection scanning, a non-linear amplifier for gamma control, beam blanking for D.C. restoration, and a separate monitor for photographic recording. Cathodoluminescent images were also obtained. A photograph of the microscope as it was in 1953 is shown in Fig. 3.

Fig. 3. Photograph of Cambridge SEM1 taken in 1953. [38, p.52].

4. Improvements to the SEM

When McMullan left Cambridge in 1953, the microscope was taken over by K.C.A. Smith [18] who made substantial improvements including more efficient collection of secondary electrons, lens stigmation, and a completely new, easily operated, specimen chamber with a rotating and tilting stage (Fig. 4). By 1955 many different specimens had been examined [19] and he had demonstrated the importance of the forward and backscattered low-energy secondary electrons (FSE, BSE) using the Baxter electron multiplier with improved electronics. But it was very bulky and, at the suggestion of Oatley, Smith tried the combination of a plastic scintillator and photomultiplier; later developed by Everhart and Thornley as the detector that is still the standard type for SEMs [7].

The selection of suitable specimens to demonstrate the capabilities of the new SEM was not easy. Examination of samples that could be imaged in a TEM either directly in reflection at a grazing angle or with a replica, was hardly likely to convince a skeptical audience of the merits of the SEM, owing to its inferior resolution at that time. However, some initial studies of surfaces of specimens that presented difficulties for the replica or reflection techniques were undertaken. These included the surfaces of wool and artificial fibres, the surface of a mealworm grub, the edge of a razor blade and the tip of a gramophone needle. The ease and simplicity with which such specimens could be prepared and examined in comparison to replicas was a revelation to those involved. Oatley then suggested [19] an application that would be unquestionably a convincing demonstration of the capabilities of the SEM: this was the imaging of a tungsten/germanium point-contact rectifier (Fig. 5) and a study of the forming processes used in its manufacture. It was obvious that replicas were inapplicable, and that with the TEM used in reflection the region of interest in the vicinity of the point contact would be completely obscured by the tungsten whisker itself. Semiconductor manufacturers with whom Oatley had extensive contacts supported the research.

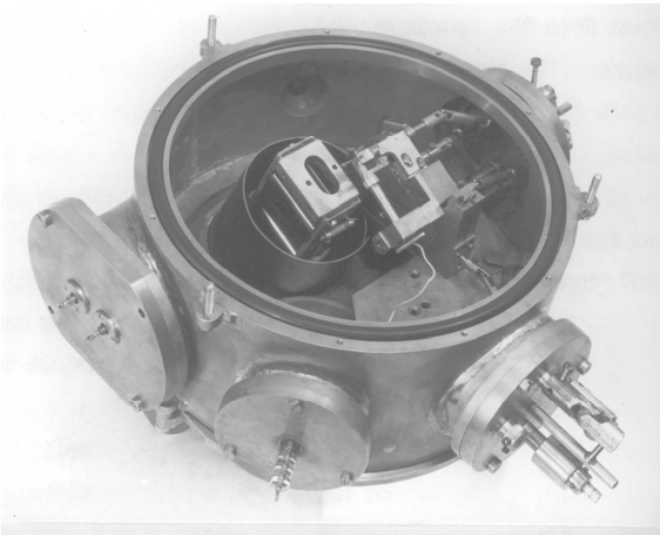

Fig. 4. Improved specimen facilities and electron collection arrangement in the second phase of SEM1 development, 1956. [18]

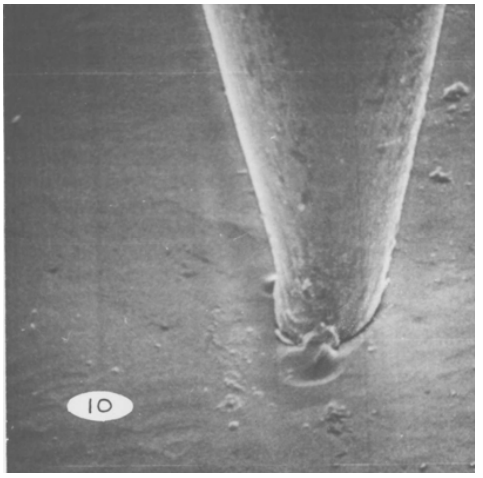

Fig. 5. SEM1: tungsten whisker placed in contact with the surface of a crystal of germanium. [19]

A micromanipulator was constructed which, mounted on the tilting and rotating stage, allowed the tungsten point to be lowered to the germanium surface at a selected location, and the pressure on the point increased in controlled increments. The 'forming' process, which was performed while the sample was under observation in the SEM, involved the application of pulses of current to produce the desired rectifier characteristics, these being monitored dynamically on a CRT display. It was thus possible for the first time to observe at high resolution the changes occurring in the region of the tip as a result of forming, and to lift the point from the surface and examine the cratered region remaining (Fig. 5 demonstrates that melting of the germanium has occurred). It was also possible to probe the surrounding region with a fresh whisker and to measure the electrical characteristics at increasing distances from the original contact point.

It was unfortunate that during the course of these experiments, research interest in the semiconductor industry switched almost entirely to junction devices, consequently the work was terminated before being fully completed and was not submitted for publication.

It is perhaps worth recording that during these experiments it was observed that variation of the bias voltage on the rectifier resulted in variations of image intensity across the field of view. What Smith failed to appreciate at the time was that he was witnessing the phenomenon later termed by Oatley and Everhart as ‘voltage contrast’ [20], which was to play such a large part in the future evolution of the SEM.

5. First outside users

In 1955 some of the first samples representing industrial and academic materials research interests, and which resulted in collaborative publications in the following year, arrived from outside the CUED. One of the first [21] was brought to the Department by J.W. Allen who was then employed by Ericsson Telephones, Nottingham. He was studying the features observed on the etched surfaces of zone-grown germanium, that indicated the emergence of edge dislocations associated with a space-charge region. The reduced action of the etch in such regions gave rise to ‘tumps’ raised from the surrounding area. Ease of specimen preparation coupled with unambiguous interpretation of images revealed the SEM to be greatly superior to the replicas employed hitherto for this work.

In addition to the regular arrays of humps seen along the line of dislocations, spectacular crystallographic features were discovered on these surfaces that had not previously been observed (Fig. 6). These would have been difficult or impossible to study using standard replica techniques.

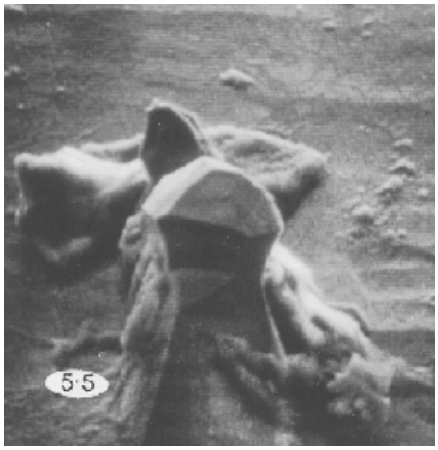

Fig. 6. SEM1: a feature on the etched surface of a zone-grown single crystal of germanium. [21]

At about the same time in the University Department of Physical Chemistry, under F.P. Bowden, attempts were being made to use a TEM at grazing angles to study the slow thermal decomposition of silver azide crystals (2AgN3 → 2Ag + 3N2). These had failed owing to the premature ignition of the crystals under the intense electron bombardment necessary in this mode of operation. The SEM appeared to be suitable for this application because its intensity of bombardment was lower by several orders of magnitude, its angle of observation was 25-30 degrees from the surface as compared with 5 degrees in the reflection TEM resulting in less foreshortening in the image and masking of detail in the foreground, and it had potentially greater possibilities regarding the dynamic observation of the decomposition process. These advantages were recognised by J. McAuslan who had been seconded by I.C.I. Nobel Division to work with Bowden on this material, and he supplied samples for examination in the SEM.

After some initial trials verifying that the azide could be examined in the SEM without initiating decomposition, it was decided to conduct in situ experiments by depositing crystals of azide directly from solution onto a small disk of silver, which was then mounted on a heater assembly held in the chuck of the specimen stage. The temperature of the plate, measured by means of a thermocouple, could be raised to a maximum of 400°C. By increasing the temperature slowly in controlled increments, the decomposition process could be readily followed. Both azide platelets and needles were studied in this way [22], complete decomposition occurring after periods extending up to 8 hours, which made heavy demands on the stability of the microscope. Some of the most informative results were obtained from the study of needles of azide that had been deposited with one end in close thermal contact with the hot plate. In such cases decomposition proceeded from this end, and it was possible to obtain micrographs at various stages of the process as the decomposition zone travelled along the needle (Fig. 7). A montage of these micrographs proved to be an informative way of displaying the results of an experiment.

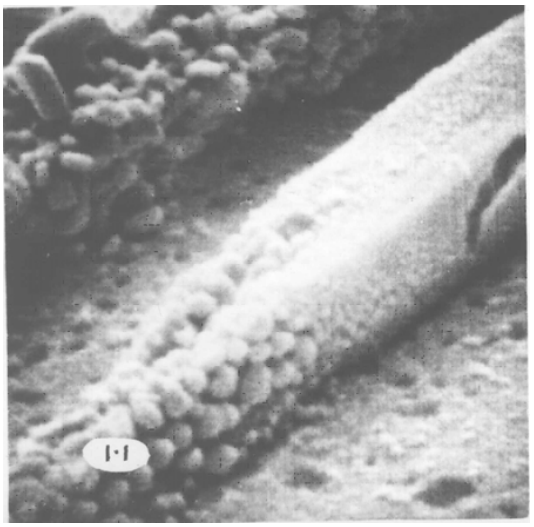

Fig. 7. SEM1: micrograph of a partly decomposed needle of silver azide. [22]

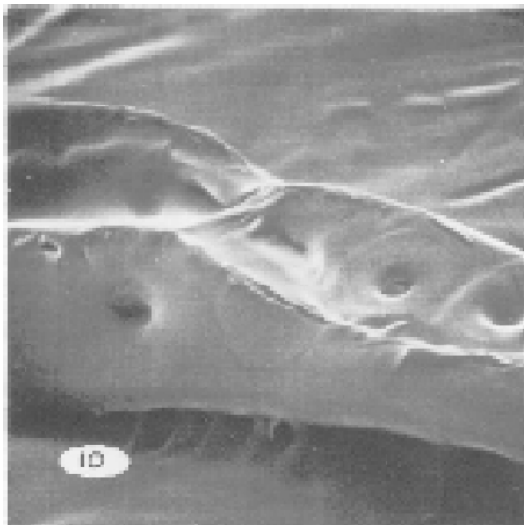

Fig. 8. SEM1: micrograph of a portion of an intact summerwood spruce fiber. [23]

Also through the assistance of the Department of Physical Chemistry, a visitor, D. Atack [23] on leave from the Pulp and Paper Research Institute of Canada (PPRIC), had samples of wood fibre that could only be examined in a TEM with great difficulty using replicas. The nature of the surfaces rendered the interpretation of the results extremely complicated. For the initial studies using the SEM, specimens of single fibres were taken directly from a mill grinder pit and floated down on to a specimen stub, allowed to dry in air and metallised with a layer of silver. It was found that this simple procedure permitted the examination of intact fibres without collapse of the outer membrane (Fig. 8). The studies were later extended to the surfaces of chemical fibres, woodchips, and newsprint.

The President of PPRIC, L.R. Thiesmeyer, was impressed by the results obtained with the SEM and, since no commercial scanning instruments were available, he ordered a microscope from Oatley and CUED. This instrument, SEM3, the first in the CUED to incorporate magnetic lenses and a high-quality display cathode-ray tube, was designed by Smith and utilized components from a surplus TEM manufactured by the AEI Company [24]. It was installed by Smith in the Montreal laboratories of the PPRIC in 1958, and he stayed there for a further two years to help with the application of the instrument to a wide variety of problems in the pulp and paper industry. It was used for several years until it was replaced by one of the first commercially manufactured instruments.

In 1953 O.C. Wells [25] joined Oatley's laboratory and was given the task of building a new instrument, SEM2, similar to the earlier one but incorporating improvements that had been shown to be desirable. He was greatly interested by McMullan's comments [14] on the imaging of surfaces using the faster scattered electrons and carried out experiments using BSE and FSE detectors in different positions to subtend large solid angles at the surface of the specimen. This line of work is still producing original results [26].

In his last year Wells used his SEM to examine a number of different specimens. This included a collaboration with Constance Tipper, a lecturer in the CUED, to obtain stereoscopic pairs of micrographs. Tipper was well known to metallurgists as the inventor of the 'Tipper Test' for determining the brittleness of steel used in the construction of ships; this had been of great importance during the Second World War. In 1956 she was breaking single crystals of iron at liquid nitrogen temperature to produce flat fracture surfaces with asperities and cavities of various kinds. These were ideal samples for showing the way in which the SEM can be used to examine and compare the corresponding features on two opposing surfaces (Fig. 9). The geometry of each half of the fracture was calculated from pairs of images taken from two directions at right angles [27]. Tipper thus became the earliest metallurgist to use the SEM for the study of fractures.

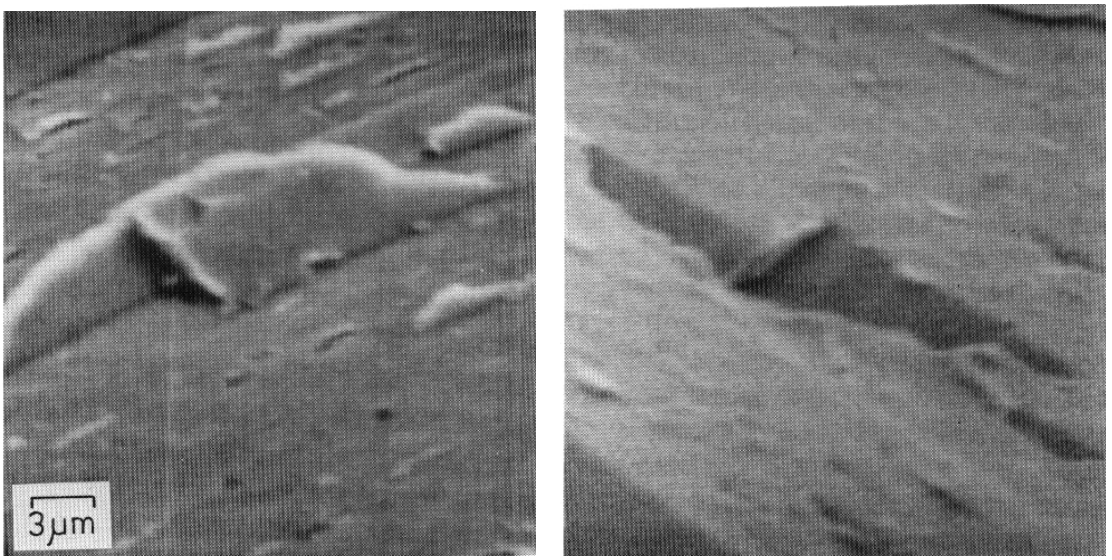

Fig. 9. SEM2: micrograph of two mating halves of fracture surfaces of a single crystal of α-iron. [27]

6. Further SEM developments at CUED

Following Smith and Wells a succession of students worked both on the instrumental development of the SEM and its applications. Advances made in Oatley's group up to 1966 include voltage contrast [20], low voltage (1 kV) SEM [28], high temperature specimens (thermionic cathodes) [29], high resolution SEM [30, 31], etching of surfaces [32], ion etching and microfabrication [33], and microelectronics [34]. Some of the historical background to this and later work in Oatley's group, and elsewhere, is described in Refs [35-38].

7. Commercial development of the SEM

In 1960 Oatley hoped that following Smith's success with SEM3, the A.E.I. Company Ltd might manufacture and market SEMs for which it was becoming clear that there would be a demand, but the instrument they developed proved to be a disappointment and he therefore looked for another firm [11]. By this time the Cambridge Instrument Company (CIC) was marketing the Microscan electron-probe X-ray microanalyser that had been developed from the original scanning electron microanalyser, which had been started in V.E. Cosslett's group at the Cavendish Laboratory by P. Duncumb and then further developed by D.A. Melford at Tube Investments Research Laboratories near Cambridge. As CIC were looking for another electron optical product and the SEM was the obvious candidate, Oatley was able to terminate the AEI agreement and negotiate a new one with Cambridge Instruments. The only outstanding problem was the transfer of the technical know-how, but this again was solved in the best possible way: one of Oatley's Ph.D. students, A.D.G. Stewart, was at the end of his course and was taken on by the Company to develop the SEM, which they marketed as the "Stereoscan" in 1965 (Fig. 10).

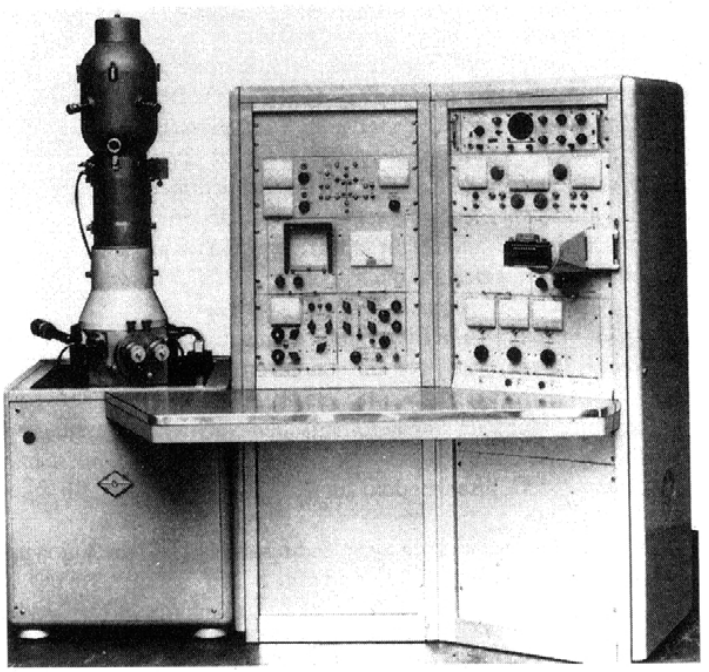

Fig. 10. Cambridge Instrument Company: prototype of the first Stereoscan SEM, 1965. [38, p.32].

Finally mention must be made of the SEM developments in other laboratories that were taking place during the period, as evidenced in scientific publications. Apart from AEI there were two other groups. A SEM was built in France in 1956 at the National Institute of Applied Science in Lyon; it had a probe size of the order of a micrometre and was used mainly for cathodoluminescence studies over a period of several years [36]. And in the USSR there was a SEM at Moscow University [37] from about 1960. No doubt there were other groups who did not publish at the time, in particular JEOL, which marketed a SEM about 6 months after the Stereoscan, following a report by one of their engineers, Y. Nogiuchi, who had seen SEM3 in operation at the Pulp and Paper Research Institute in Montreal [38, p. 541].

8. Conclusion

It is clear that the years 1950 - 1965 were of the greatest significance for the original development of the SEM and the scanning microprobe analyser and that the major part of this work was done in the four Cambridge institutions that have been mentioned: namely, the Cambridge University Engineering Department, the Cavendish Laboratory, the Tube Investment Research Laboratories, and the Cambridge Instrument Company. However, all this would not have happened in Cambridge without two men, Sir Charles Oatley who was the driving force towards the goal of scanning electron microscopy, and Ellis Cosslett, who saw clearly the advantages of scanning in microprobe X-ray analysis. Their contributions in these fields are recorded in a recently published volume [38] written by many of those who worked under them and benefited from their inspiring leadership. Without them it would seem that the introduction of these instruments might have been delayed for several more years.

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